

08/894351

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

INTERNATIONAL APPLICATION NO. PCT/EP96/00605	INTERNATIONAL FILING DATE FEBRUARY 13, 1996	PRIORITY DATE CLAIMED FEBRUARY 18, 1995
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TITLE OF INVENTION  
GAS-PRODUCING MIXTURES

APPLICANT(S) FOR DO/EO/US

Klaus REDECKER, Waldemar WEUTER, Ulrich BLEY

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1.  This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
2.  This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
3.  This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4.  A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5.  A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a.  is transmitted herewith (required only if not transmitted by the International Bureau).
  - b.  has been transmitted by the International Bureau.
  - c.  is not required, as the application was filed in the United States Receiving Office (RO/US).
6.  A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7.  Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a.  are transmitted herewith (required only if not transmitted by the International Bureau).
  - b.  have been transmitted by the International Bureau.
  - c.  have not been made; however, the time limit for making such amendments has NOT expired.
  - d.  have not been made and will not be made.
8.  A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9.  An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10.  A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11.  An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12.  An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13.  A FIRST preliminary amendment.
- A SECOND or SUBSEQUENT preliminary amendment.
14.  A substitute specification.
15.  A change of power of attorney and/or address letter.
16.  Other items or information:
  - \*\* INTERNATIONAL PUBLICATION NO. WO96/26169
  - \*\* PCT REQUEST FORM
  - \*\* INTERNATIONAL PRELIMINARY EXAMINATION REPORT

17.  The following fees are submitted:

## BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5) ):

Search Report has been prepared by the EPO or JPO ..... \$910.00

International preliminary examination fee paid to USPTO (37 CFR 1.482) ..... \$700.00

No international preliminary examination fee paid to USPTO (37 CFR 1.482)  
but international search fee paid to USPTO (37 CFR 1.445(a)(2)) ..... \$700.00Neither international preliminary examination fee (37 CFR 1.482) nor  
international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$1,040.00International preliminary examination fee paid to USPTO (37 CFR 1.482)  
and all claims satisfied provisions of PCT Article 33(2)-(4) ..... \$96.00

ENTER APPROPRIATE BASIC FEE AMOUNT =

\$ 910.00

Surcharge of \$130.00 for furnishing the oath or declaration later than  20  30  
months from the earliest claimed priority date (37 CFR 1.492(e)).

\$

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	24	- 20 =	X \$22.00	\$ 110.00
Independent claims	4	- 3 =	X\$80.00	\$ 80.00
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+\$260.00	\$

TOTAL OF ABOVE CALCULATIONS = \$ 1,100.00

Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement  
must also be filed (Note 37 CFR 1.9, 1.27, 1.28).

\$

SUBTOTAL = \$ 1,100.00

Processing fee of \$130.00 for furnishing the English translation later than  20  30  
months from the earliest claimed priority date (37 CFR 1.492(f)).

\$

TOTAL NATIONAL FEE = \$ 1,100.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be  
accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property

\$

TOTAL FEES ENCLOSED = \$ 1,100.00

Amount to be:  
refunded  
charged \$a.  A check in the amount of \$ 1,100.00 to cover the above fees is enclosed.b.  Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \$ \_\_\_\_\_ to cover the above fees.  
A duplicate copy of this sheet is enclosed.c.  The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any  
overpayment to Deposit Account No. 01-2135. A duplicate copy of this sheet is enclosed.NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR  
1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

ANTONELLI, TERRY, STOUT & KRAUS  
1300 NORTH SEVENTEENTH STREET  
SUITE 1800  
ARLINGTON, VA 22209

SIGNATURE:

ALAN E. SCHIAVELLI

NAME

32,087

REGISTRATION NUMBER

306.35565X00

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: REDECKER et al

Serial No.:

Filed: August 18, 1997

For: GAS PRODUCING MIXTURES

Group:

Examiner:

**PRELIMINARY AMENDMENT**Assistant Commissioner  
for Patents  
Washington, D.C. 20231

August 18, 1997

Sir:

Prior to examination on the merits of this application and prior to  
calculation of the filing fee, please amend the above-identified application  
as follows:

**IN THE CLAIMS:**

Claim 4, line 1, delete "any one of claims" and insert --Claim--;  
line 2, delete "to 3".

Claim 5, line 1, delete "any one of claims" and insert --Claim--;  
line 5, delete "to 4".

Claim 6, line 1, delete "any one of claims" and insert --Claim--;  
line 2, delete "to 5".

Claim 7, line 1, delete "any one of claims" and insert --Claim--;  
line 2, delete "to 6".

Claim 8, line 1, delete "any one of claims" and insert --Claim--;  
line 2, delete "to 7".

Claim 9, line 1, delete "any one of claims" and insert --Claim--;  
line 2, delete "to 8".

Claim 10, line 1, delete "any one of claims" and insert --Claim--;  
line 2, delete "to 9".

Claim 11, line 1, delete "any one of claims" and insert --Claim--;  
line 2, delete "to 10".

Claim 12, line 1, delete "any one of claims" and insert --Claim--;  
line 2, delete "to 11".

Claim 13, line 1, delete "any one of claims" and insert --Claim--;  
line 2, delete "to 12".

Claim 14, line 1, delete "any one of claims" and insert --Claim--;  
line 2, delete "to 13".

Claim 15, line 1, delete "any one of claims";  
line 2, delete "1 to" and insert --Claim--.

18. (Amended) Gas-producing agent according to [claim 16 or] claim 17, characterised in that [the] it contains as oxidants [are defined according to claim 6] and [the] contains as combustion moderators [are defined according to any one of claims 10 to 12]:

peroxides of alkali and alkaline earth metals, zinc peroxide, and peroxodisulphates of the said elements and ammonium peroxodisulphate, or mixtures of these compounds;

ammonium nitrate, nitrates of alkali and alkaline earth metals, in particular lithium nitrate, or mixtures of these compounds;

halogen oxycompounds of alkali or alkaline earth metals or of ammonium, preferably potassium perchlorate or ammonium perchlorate, or mixtures of these compounds, substances or mixtures thereof which are capable of influencing the combustion and its rate by heterogeneous or homogeneous catalysis, the proportion of these substances in the mixture amount up to 8%.

Claim 22, line 1, delete "any one of claims" and insert --Claim--;  
line 1, delete "to 21".

Claim 23, line 2, delete "any one of claims" and insert --Claim--;  
line 2, delete "to 18".

Claim 24, line 2, delete "any one of claims" and insert --Claim--;  
line 2, delete "to 18".

REMARKS

The foregoing amendments are respectfully requested prior to examination on the merits of this application.

To the extent necessary, applicants petition for an extension of time under 37 CFR 1.136. Please charge any shortage in the fees due in connection with the filing of this paper, including extension of time fees, to the deposit account of Antonelli, Terry, Stout & Kraus, LLP, Deposit Account No. 01-2135 (Case: 306.35565X00), and please credit any excess fees to such deposit account.

Respectfully submitted,

ANTONELLI, TERRY, STOUT & KRAUS, LLP



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Alan E. Schiavelli  
Registration No. 32,087

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306.35565X00

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: REDECKER et al

Serial No.:

Filed: August 18, 1997

For: GAS PRODUCING MIXTURES

Group:

Examiner:

**SECOND PRELIMINARY AMENDMENT**Assistant Commissioner  
for Patents  
Washington, D.C. 20231

August 18, 1997

Sir:

Prior to examination on the merits of this application, please amend  
the above-identified application as follows:

**IN THE CLAIMS:**

Please add the following new claim to the application:

--25. Gas-producing agent according to claim 16, characterised in that it  
contains as oxidants and contains as combustion moderators:

peroxides of alkali and alkaline earth metals, zinc peroxide, and  
peroxodisulphates of the said elements and ammonium peroxodisulphate,  
or mixtures of these compounds;

ammonium nitrate, nitrates of alkali and alkaline earth metals, in

particular lithium nitrate, or mixtures of these compounds; halogen oxycompounds of alkali or alkaline earth metals or of ammonium, preferably potassium perchlorate or ammonium perchlorate, or mixtures of these compounds, substances or mixtures thereof which are capable of influencing the combustion and its rate by heterogeneous or homogeneous catalysis, the proportion of these substances in the mixture amount up to 8%.

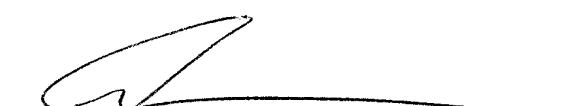
REMARKS

The foregoing amendments are respectfully requested prior to examination on the merits of this application.

To the extent necessary, applicants petition for an extension of time under 37 CFR 1.136. Please charge any shortage in the fees due in connection with the filing of this paper, including extension of time fees, to the deposit account of Antonelli, Terry, Stout & Kraus, LLP, Deposit Account No. 01-2135 (Case: 306.35565X00), and please credit any excess fees to such deposit account.

Respectfully submitted,

ANTONELLI, TERRY, STOUT & KRAUS, LLP



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"Gas-producing mixtures"

Gas generators are being used to an increasing extent, for example in motor vehicles for life-saving purposes. The gas-producing mixture usually contains sodium azide. Sodium azide as such is poisonous, and it can readily react with heavy metals, e.g. copper and lead, to form extremely dangerous and vigorously reacting compounds. Special precautions must therefore be taken in the production of the raw material and of the gas charge mixture, in its processing and in quality control. For this reason the disposal of the sodium azide, for example when exchanging defective gas generators or when scrapping vehicles, also presents a particular problem. Improper use must also be reliably prevented.

There has been no lack of attempts to use other substances in place of sodium azide. A common feature of all proposed replacements for sodium azide is that they contain organic carbon compounds, and as a rule also organic nitrogen compounds. EP 0 519 485 describes the use of tetrazole or a derivative or derivatives of tetrazole, or the use of one or more compounds from the group consisting of cyanic acid derivatives and their salts, one or more compounds from the group consisting of triazine and triazine derivatives, the use of urea, its salts, derivatives and salts of these compounds: these compounds can also be present as mixtures. Ammonium nitrate and nitrates of sodium, potassium, magnesium, calcium and iron, and/or peroxides of zinc, calcium, strontium or magnesium, can be used as oxidants. Other gas-producing components, cooling agents, reducing agents, catalysts and/or porosity producing agents can be added.

EP 0 438 851 describes a non-toxic, non-azide

pyrotechnic composition which is suitable for use in the production of substantially non-toxic combustion products that include a gas in order to fill an accident cushion. The composition includes a mixture 5 of at least one tetrazole or tetrazole compound containing hydrogen in its molecule, at least one oxygen-containing oxidant and at least one metal oxide selected from cobalt oxide, nickel oxide, chromium oxide, aluminium oxide and boron oxide. In the 10 combustion a substantially non-toxic primary gas mixture and filterable solids are produced. Aminotetrazole, together with oxidants which can contain perchlorates as well as nitrates, is employed, by way of example.

15 A similar formulation is disclosed in European patent EP 0 372 733: the use of tetrazoles and triazoles in mixtures with ammonium perchlorate and alkali nitrate as oxidants, in combination with an additive to control combustion.

20 PCT application WO 94/01381 describes a gas-producing agent for airbags consisting of organic nitrocompounds and halogenates. By halogenates are to be understood, for example, alkali metal chlorates, bromates and their per-compounds. The following are 25 mentioned, *inter alia*, as combustion-controlling catalysts: oxides, chlorides, carbonates, sulphonates of the 4th to 6th series of the periodic classification.

When the above-mentioned gas charges undergo 30 reaction to inflate airbags for motor vehicle safety, proportions of toxic gases such as, for example, carbon monoxide or nitrogen oxides can be present besides non-toxic working gases such as nitrogen, carbon dioxide and hydrogen. Limits such as, for example, the maximum 35 allowable concentration (MAC) in the workplace, are set having regard to peak loads for these gases. Their

formation is thermodynamically and kinetically connected, and in the case of carbon monoxide is controlled e.g. by the producer gas equilibrium. It has further been found that mixtures which contain 5 nitrogen- and carbon-containing compounds and evolve small proportions of  $\text{NO}_x$  during combustion evolve large proportions of CO, and vice versa. The establishment of these equilibria is temperature and pressure-dependent. It is known that a sufficiently effective 10 influencing of the composition of the reaction gases towards the formation of non-toxic products cannot be achieved solely by physical measures, for example control of the reaction by pressure and/or temperature.

Processes are known from the literature which have 15 the object of reducing these components of the reaction gases. Thus, for example, the equilibrium can be displaced at the expense of carbon monoxide formation by the formation of carbonates by the addition of alkaline slag-forming agents to the gas charge. At the 20 same time nitrogen oxides are converted into nitrates or nitrites.

However, these measures have the disadvantage that the yield of gas is made substantially worse by the high proportion of slag. In addition, the slag must be 25 separated at some expense from the gaseous constituents by filters or other retaining systems before the working gases can be used, for example for the inflation of the airbag.

While the use of nitrogen-free systems does lead 30 to the formation of nitrogen-free gases, this is at the expense of a lower yield of gas. The reason for this is that, to displace the producer gas equilibrium in the direction of  $\text{CO}_2$ , an excess of slag-forming oxygen-carrying agent must be used. Hence hybrid systems have 35 already been proposed in which the reactions described above are brought about by compressed air instead of by

slag-forming oxidants. However, these concepts suffer from the disadvantage of the high weight of the system and the need to control or supplement the compressed air.

5 According to US patent 3 910 595, to improve the yield the gas forming in the reaction is passed through a venturi nozzle so that ambient air can be drawn in to assist in the inflation of the airbag. Here, however, it must be taken into account that this ambient air  
10 greatly cools the hot gases. Particularly at low ambient temperatures the resulting loss in volume for inflating the gasbag must be compensated for by the pyrotechnic mixture. The resulting increased proportion of toxic reaction gases in the interior of  
15 the vehicle can no longer be sufficiently reduced by dilution.

The present invention provides non-toxic, azide-free mixtures for the production of gas by combustion. These gas-producing mixtures can be used, *inter alia*,  
20 in safety devices, for example in airbag systems for inflation of airbags in motor vehicles and aircraft. However, they are also suitable for lifting heavy loads by inflation of bags placed under them, or for  
25 expulsion of e.g. fire extinguishing powder, or for other measures where the performance of work requires rapid formation of gases.

The mixtures in accordance with the invention contain:

- a) as nitrogen-containing compound (fuel) at least one compound from the group: tetrazole, triazole, triazine, cyanic acid, urea, their derivatives or their salts;
- b) as oxidant, at least three compounds from the group of the peroxides, nitrates, chlorates or perchlorates;
- c) combustion moderators which are capable of

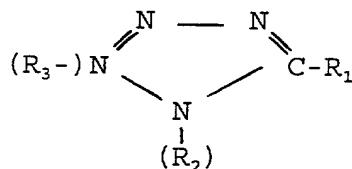
influencing the combustion and its rate by heterogeneous or homogeneous catalysis; and optionally also

5 d) additions which are capable of reducing the proportion of the toxic gases.

The mixtures in accordance with the invention are not toxic, and in contrast to azide-containing mixtures are easy to handle. They therefore require less outlay on safety in the production of the raw materials and  
10 mixtures and in their shaping, storage or disposal.

The nitrogen-containing compounds to be used according to the invention are those which, in the mixture with oxidants, mainly form in their thermal/chemical reaction  $\text{CO}_2$ ,  $\text{N}_2$ ,  $\text{O}_2$  and  $\text{H}_2\text{O}$ , but do not  
15 evolve any gases such as  $\text{CO}$  or  $\text{NO}_x$  in concentrations that could endanger health.

The mixtures according to the invention preferably contain as nitrogen-containing compounds (fuels) one or more tetrazole derivatives of the formula:  
20



25 in which  $\text{R}_1$  and  $\text{R}_2$  or  $\text{R}_3$  can be the same or different, with either  $\text{R}_2$  or  $\text{R}_3$  being present, and standing for hydrogen, hydroxy, amino, carboxyl, an alkyl radical with 1 to 7 carbon atoms, an alkenyl radical with 2 to  
30 7 carbon atoms, an alkylamino radical with 1 to 10 carbon atoms, an aryl radical, optionally substituted with one or more substituents which can be the same or different and are selected from the amino group, the nitro group, alkyl radicals with 1 to 4 carbon atoms or  
35 an arylamino radical in which the aryl radical can optionally be substituted, or the sodium, potassium and

guanidinium salts of the said tetrazole derivatives.

In these compounds:

$R_1$  preferably stands for hydrogen, amino, hydroxy, carboxyl, a methyl, ethyl, propyl or isopropyl, butyl, isobutyl or tert-butyl, n-pentyl, n-hexyl, or n-heptyl radical, a methylamino, ethylamino, dimethylamino, n-heptylamino, n-octylamino or n-decylamino radical, a tetrazole radical, a phenylamino radical, a phenyl, nitrophenyl or aminophenyl radical; and

10 R<sub>2</sub> or R<sub>3</sub> preferably stands for hydrogen, a methyl or ethyl radical, a phenyl, nitrophenyl or aminophenyl radical.

Particularly preferred compounds are the tetrazole derivatives 5-aminotetrazole, lithium, sodium,

15 potassium, zinc, magnesium, strontium or calcium 5-aminotetrazolate, 5-aminotetrazole nitrate, sulphate, perchlorate and similar compounds, 1-(4-aminophenyl)-tetrazole, 1-(4-nitrophenyl)-tetrazole, 1-methyl-5-dimethylaminotetrazole, 1-methyl-5-methylamino  
20 tetrazole, 1-methyltetrazole, 1-phenyl-5-aminotetrazole, 1-phenyl-5-hydroxytetrazole, 1-phenyltetrazole, 2-ethyl-5-aminotetrazole, 2-methyl-5-aminotetrazole, 2-methyl-5-carboxytetrazole, 2-methyl-5-methylaminotetrazole, 2-methyltetrazole, 2-  
25 phenyltetrazole, 5-(p-tolyl)tetrazole, 5-diallylmino tetrazole, 5-dimethylaminotetrazole, 5-ethylamino tetrazole, 5-hydroxytetrazole, 5-methyltetrazole, 5-methylaminotetrazole, 5-n-decylaminotetrazole, 5-n-heptylaminotetrazole, 5-n-octylaminotetrazole, 5-  
30 phenyltetrazole, 5-phenylaminotetrazole or bis-(aminoguanidine)-azotetrazole and diguanidinium-5,5'-azotetrazolate, as well as 5,5'-bitetrazole and its salts, such as the 5,5'-bi-1H-tetrazole ammonium compounds.

35 The mixtures may contain: as triazine derivatives,  
1,3,5-triazine, as triazole derivatives, 1,2,4-

triazole-5-one, 3-nitro-1,2,4-triazole-5-one, as cyanic acid derivatives, sodium cyanate, cyanuric acid, cyanuric acid esters, cyanuric acid amide (melamine), 1-cyanoguanidine, sodium dicyanamide, disodium 5 cyanamide, dicyanodiamidine nitrate, dicyanodiamidine sulphate, and as urea derivatives biuret, guanidine, nitroguanidine, guanidine nitrate, aminoguanidine, aminoguanidine nitrate, thiourea; triaminoguanidine nitrate, aminoguanidine hydrogen carbonate, 10 azodicarbonamide, tetracene, semicarbazide nitrate, as well as urethanes, ureides such as barbituric acid, and derivatives thereof.

5-aminotetrazole is used as a particularly preferred component. When this component is used in 15 the mixture the preferred proportion is 10-40% by wt. As derivatives of 5-aminotetrazole, its salts in which the acidic hydrogen atoms in 5-aminotetrazole are replaced in salt-like manner by toxicologically acceptable elements such as calcium, magnesium or zinc, 20 are used. However, compounds in which the cation is ammonium, guanidinium and its amino derivatives can also be used.

Oxidants which may be used according to the invention are:

25 - peroxides of alkali and alkaline earth metals, zinc peroxide, and the peroxodisulphates of the said elements and ammonium peroxodisulphate;

- ammonium nitrate, nitrates of alkali and alkaline earth metals, in particular lithium, sodium or potassium nitrate, and strontium nitrate;

- halogen oxycompounds of the alkali or alkaline earth metals or of ammonium, 30 particularly preferably potassium perchlorate or ammonium perchlorate.

The oxidants can be used singly or in mixtures. In order to reduce the proportion of nitrogen oxides in the reaction mixture as far as possible, it is advantageous to keep the proportion of nitrate in the 5 oxidant mixture as small as possible, since part of the nitrate can undergo thermal decomposition.

A preferred combination of the oxidants consists of zinc peroxide, potassium perchlorate and at least one nitrate, preferably sodium nitrate or strontium 10 nitrate, mixed in the ratio 1:2:10 and in a total amount of about 60% by wt. in the gas-producing mixture. The chlorine-containing compounds then react during the combustion to form harmless sodium/potassium chloride. Ammonium perchlorate can also be considered 15 as perchlorate, alone or mixed with another halogen oxycompound, but an excess must be avoided in order to prevent the formation of corrosive hydrochloric acid. If ammonium perchlorate is used, the simultaneous presence of zinc compounds is particularly 20 advantageous, since the risk of hydrochloric acid being formed can thereby be avoided. An excess of sodium and potassium compounds is acceptable, since these compounds react with the reaction gases to form harmless carbonates. The partial or complete 25 replacement of the alkali nitrate by strontium nitrate leads to a marked reduction in the amount of slag.

The ratio of the nitrogen-containing compounds, for example the tetrazoles and triazoles, to the oxidants in the mixture is balanced so that, on 30 combustion of the gas charge mixture, an excess of oxygen is formed. This excess of oxygen displaces the CO/CO<sub>2</sub> equilibrium in the direction of carbon dioxide.

Substances or mixtures thereof which are capable by heterogeneous or homogeneous catalysis of 35 influencing the combustion and its rate are used as combustion moderators. Moderators which intervene in

the reaction through heterogeneous catalysis are metals, metal oxides and/or metal carbonates and/or metal sulphides. Preferred metals to use are boron, silicon, copper, iron, titanium, zinc or molybdenum.

5 Calcium carbonate can also be used. Mixtures of these moderators can likewise be used.

Moderators which intervene in the reaction through homogeneous catalysis are, for example, sulphur, boron, silicon or ferrocene and its derivatives. These

10 moderators are vapourized into the vapour phase as a result of the temperatures occurring in the reaction, and thus can intervene in the reaction either as such or as after-products. The proportion of these substances in the mixture can amount to up to about 8%

15 Furthermore the mixture according to the invention can contain gas-producing additions which are capable of reducing the proportion of the noxious gases such as nitrogen oxides and/or carbon monoxide. The proportion of these noxious gases in the gas mixture produced is

20 determined by

- the stoichiometric composition of the mixture,
- the temperature and pressure of the reaction,
- additives for influencing the reaction or the after-reaction, and by the
- 25 - design of the generator in which the reaction takes place.

While in a closed system, such as, for example, a pressure bomb, it is relatively easy to reach the composition of the gas mixture which approximates to 30 the thermodynamic calculations, this can no longer be achieved under the actual operating conditions in the generator, since during the few milliseconds duration of the reaction the equilibrium cannot be established. According to the invention, therefore, suitable 35 substances which can bring about a catalytic effect are introduced into the mixture or into the region of the

outflowing gases. For this purpose the combustion moderators described above and oxides of precious metals can be used. Further possibilities consist in the use of noble metals such as palladium, ruthenium, 5 rhenium, platinum or rhodium, which employ the excess oxygen in the reaction gases in a subsequent reaction to convert the carbon monoxide. A preferred mode of use contemplates applying the additive materials on ceramic or electrodepositing them on metal grids as a 10 support. Using this method it is possible in particular to reduce the proportion of carbon monoxide in the gas mixture.

To reduce the proportion of  $\text{NO}_x$ , additional substances are used whose chemical properties catalyse 15 in particular the conversion of nitrogen oxides, for example nitrogen dioxide, to nitrates or nitrites. In principle, all more or less strongly basically reacting substances are suitable.

These include, for example, oxides, hydroxides or 20 carbonates of non-toxic elements such as, for example, those of the alkali and alkaline earth metals, those of zinc, and mixtures of these compounds. When these compounds are used, nitrates and nitrites of the elements are mainly formed. Further suitable materials 25 for reaction with  $\text{NO}_2$  are urea, guanidine and its derivatives, compounds having  $\text{NH}_2$  groups, such as, for example, amidosulphonic acids, amido complexes and the like, and amides. A particularly preferred embodiment contemplates the use of peroxides in the outlet 30 openings of the generator. A particular advantage of this is that, beside the reduction of the nitrogen oxides by the reaction described above, oxygen is also formed for the following catalytic reaction with carbon monoxide.

35 The additions according to the invention, either alone or together, can either be introduced directly

into the gas-producing charge or be placed in the outflow passages of the generator. For use in the outlet passages of the generator the additions are suitably used in a compacted form, for example in the 5 form of tablets, pellets or granules. The quantity of the additions used in the charge amounts to about 10% by wt. In the outlet channels the quantity of the additions can be as much as 75% by wt., based on the gas charge.

10 A reduction in the CO content can surprisingly also be achieved if part of the fuel consists of the salts, particularly the calcium, magnesium or zinc salts, of the aminotetrazole, preferably of the corresponding salts of 5-aminotetrazole, or of urea 15 derivatives. In these cases it suffices to use only two oxidants.

To influence the rate and temperature of reaction further additives can be added. Such additives can for example be boron or metal powders, for example 20 titanium, aluminium, zirconium, iron, copper, molybdenum, as well as their stable hydrides. Their proportion in the additions is of the order of 5 % by wt.

25 The production of the gas charge mixtures according to the invention is carried out in known manner. For example, the components are mixed dry, sieved, divided into portions and pressed to tablets. The adjustment of the rate of combustion can be achieved through the shape and size of the grains of 30 the bulk material obtained by breaking and sieving out the fragments. The bulk material can be produced in large quantities and adapted to meet particular combustion requirements by mixing fractions with different dynamic liveliness. To improve the safety or 35 improve the results of mixing, premixtures of 2 or 3 components can also be used. A mixture of oxidant and

additions may, for example, be made before it comes into contact with the nitrogen-containing compounds.

However, the mixture can also be produced by kneading water-moistened components, followed by 5 granulating, e.g. by passage through sieves, extrusion or the like. In this case binders, for example waterglass, "inorganic rubber" (phosphorus chloronitrile) or even small proportions of organic binders such as acrylic resin, PTFE, or guar gum, can 10 be used. Since the components employed are neither toxic nor particularly reactive, and can only be caused to react in the enclosed space with the aid of special igniters, no special safety precautions are necessary.

The bulk material thus obtained can be used 15 directly. To avoid abrasion of the loose material in contact with the generators, which would lead to changes in the combustion characteristics and would represent a safety risk through its vigorous combustion, the bulk material can be surface-coated. 20 This can be done through a varnish coating, which can optionally be provided with ignition-promoting additions to assist in the ignition. Ignition-promoting additions that come into consideration are oxidants such as zinc peroxide and metal powders such 25 as titanium and zirconium. The application can be effected by spraying-on the solvent-containing coating agent, e.g. in a drum while evaporating off the solvent.

For special fields of application porous grain 30 structures in the grains can be used. The production of such porous structures can be effected by conventional methods, for example by adding soluble salts and subsequently dissolving them out with suitable solvents or by addition of thermally 35 decomposable substances such as, for example, ammonium bicarbonate, acetone dicarboxylic acid, blowing agents,

peroxides or azo-bisisobutyronitrile, which can then be removed again in a subsequent process step by heating and tempering at elevated temperature. The characteristic is determined by quantity, grain size 5 and distribution. Such gas charges can, for example, be used where gas charges which react in a strongly progressive manner are required.

The ignition of the tailor-made gas charge can be effected by the conventional methods. In doing so it 10 is important that no additional toxic reaction gas components are set free from the igniter after the reaction.

The gas charge mixture is insensitive in respect of its safety characteristics, for example to the 15 effects of abrasion, shock and impact or to ignitability by flame or cerium/iron sparks under normal pressure. In an enclosure, however it burns vigorously when suitably ignited. This provides increased safety in manufacture and handling.

20 The mixtures according to the invention can, for example, be used in gas generators for motor vehicle safety with the electrically initiated ignition systems conventionally employed there.

In contrast to generators based on an azide 25 charge, expensive filtering of the slag can be dispensed with, since the slag contains no toxic constituents. It consists mainly of carbonates and chlorides of potassium and sodium, along with very little nitrates/nitrites and zinc oxide. The discharge 30 of such non-toxic constituents is therefore generally only limited by the limits set for the emission of dust.

The following examples are intended to explain the invention in more detail, but without limiting it.

35 The specified components for the gas charges according to the invention are homogenised for 30

minutes in the stated proportions by weight in plastic containers in an eccentric tumbling mixer. Tabletting of the mixtures to blanks with a diameter of about 6 mm is then effected. 3.5 g of the tabletted samples are 5 caused to react using 0.2 g boron/potassium nitrate (25:75 parts by wt.) as an igniting mixture and an electrically heatable iron wire in a 25 ml stainless steel pressure bomb. The pressure-time curve of the reaction is recorded using a piezoelectric measuring 10 device. Combustion gases which are composed mainly of  $H_2O$ ,  $CO_2$ ,  $N_2$  and  $O_2$  and meet the toxicological requirements set are formed in the exothermic reaction.

The gas charge mixtures described in the examples are investigated, for example in a measuring apparatus 15 comprising a combustion chamber, gas flow diversion and filter chamber, using specific mechanical constructional conditions, to determine its combustion characteristics. The gaseous reaction products are collected and characterised in a 60 l volume vessel 20 (main constituents:  $H_2O$ ,  $CO_2$ ,  $N_2$  and  $O_2$ ).

Example No.	Composition (wt. %)					
	1	2	3	4	5	
25	5-aminotetrazole sodium nitrate potassium perchlorate	33.1 52.3 10.1	33.1 52.3 10.1	34.0 61.5 ----	33.1 52.3 10.1	34.2 64.8 ----
30	zinc oxide zinc peroxide graphite	4.0 --- 0.5	3.0 1.0 0.5	1.0 3.0 0.5	4.0 4.0 0.5	---- ---- 1.0

Example	Heat of explosion (kJ/g)	Friction sensitivity (N)	Impact sensitivity (J)
1	3.61	> 360	10
2	3.69	> 360	10
3	3.70	> 360	10
4	3.82	> 360	7.5
40	3.82	> 360	10

Results of measurements in the ballistic pressure bomb

Example	Maximum pressure (bar)	Time difference <sup>1)</sup> 40-60% p(max) (ms)	Cold gas <sup>2)</sup> (l/g)	CO (ppm)
5	1	715	6.7	0.41
	2	707	5.9	0.38
	3	729	6.1	0.41
	4	660	6.5	0.40
	5	730	6.7	0.41

10

<sup>1)</sup> duration of the reaction at between 40 and 60% of the maximum pressure, in milliseconds

15

<sup>2)</sup> measured after cooling to room temperature.

Example 1 describes the reaction of 5-aminotetrazole (5-ATZ) with a binary mixture of oxidants. The reaction gas composition shows a content of 1800 ppm CO in the reaction gases after combustion in a closed pressure bomb. In Example 2 the addition of only 1 % by wt. of zinc peroxide surprisingly leads to a marked reduction in the proportion of CO to 1100 ppm with otherwise unchanged test parameters. The changes in the composition of the mixtures in Examples 3 to 5 lead to poorer results.

30

Example No.	Composition (wt. %)				
	6 (=1)	7	8	9	
35	5-aminotetrazole	33.1	25.4	16.6	10.7
	sodium nitrate	52.3	52.7	52.7	52.7
	potassium perchlorate	10.1	10.2	10.2	10.2
	Zn (5-ATZ) <sub>2</sub>	-----	11.2	-----	-----
	Ca (5-ATZ) <sub>2</sub>	-----	-----	20.0	-----
40	Mg (5-ATZ) <sub>2</sub>	-----	-----	-----	25.9
	zinc oxide	4.0	-----	-----	-----
	graphite	0.5	0.5	0.5	0.5

45

Example		Heat of explosion (kJ/g)	Friction sensitivity (N)	Impact sensitivity (J)
5	6 (=1)	3.61	> 360	10
	7	3.64	> 360	10
	8	3.46	> 360	15
	9	2.74	> 360	20

10

Results of measurements in the ballistic pressure bomb (25 ml):

Example		Maximum pressure (bar)	Time difference <sup>1)</sup> 40-60% p(max) (ms)	Cold gas <sup>2)</sup> (l/g)	CO (ppm)
15	6 (=1)	715	6.7	0.41	1800
	7	662	6.8	0.39	250
	8	602	6.6	0.40	140
	9	81	39.2	0.33	100

20

Results of measurements in the 60 l test canister:

Example		CO reduction <sup>3)</sup> (%)	Maximum pressure <sup>4)</sup> (bar)
30	6 (=1)	0	2.2
	7	10	2.1
	8	40	1.7
	9	95	< 1.5

35      <sup>1)</sup> duration of the reaction between 40 and 60% of the maximum pressure in milliseconds

40      <sup>2)</sup> measured after cooling to room temperature

45      <sup>3)</sup> based on the test canister results, Example 1 or 6

50      <sup>4)</sup> mass of charge 40 g.

Examples 6 to 9 show that the addition of the Zn, Ca and Mg salts of 5-aminotetrazole ( $\text{Me(5-ATZ)}_2$ ) has a favourable effect on the reaction gas composition. A marked reduction is found in the proportion of CO. The rate of reaction is also affected.

		Composition (wt. %)				
		Example No.	10	11	12	13
5	5-aminotetrazole		33.0	31.6	30.8	28.9
	guanidine nitrate		8.3	8.0	7.8	7.3
	sodium nitrate		58.2	39.0	27.1	-----
	strontium nitrate		-----	20.9	33.8	63.3
	graphite		0.5	0.5	0.5	0.5

10

		Example	Heat of explosion (kJ/g)	Friction sensitivity (N)	Impact sensitivity (J)	Mass of residue <sup>3)</sup> (g)
15	10		4.06	> 360	20	1.5
	11		3.90	> 360	15	1.2
	12		3.61	> 360	20	1.0
	13		3.41	> 360	15	0.8

20

Results of measurements in the ballistic pressure bomb (25 ml):

		Example	Maximum pressure (bar)	Time difference <sup>1)</sup> 40-60% p(max) (ms)	Cold gas <sup>2)</sup> (l/g)
25	10		779	6.1	0.46
	11		767	7.0	0.41
	12		723	7.3	0.42
	13		620	8.6	0.39

30

<sup>1)</sup> duration of reaction at between 40 and 60% of the maximum pressure, in milliseconds

<sup>2)</sup> measured after cooling to room temperature

35

<sup>3)</sup> mass of solids in the 60 l test canister after combustion of 30 g gas charge in the experimental generator.

40

Examples 10 to 13 differ in the proportion of sodium nitrate/strontium nitrate used as oxidant. With increasing proportions of strontium nitrate, the mass of the slag emerging into the canister decreases. This means that the filterability of the slag is improved by

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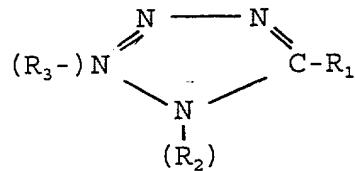
the addition of strontium nitrate - after the reaction - to the filter of the generator. At the

same time the proportion of CO in the reaction gas can be favourably influenced.

Claims

1. Gas-producing agent for gas generators, comprising nitrogen-containing compounds, characterised in that it contains:
  - 5 a) as nitrogen-containing compound (fuel), at least one compound from the group: tetrazole, triazole, triazine, cyanic acid, urea, derivatives thereof or their salts;
  - b) as oxidant, at least three compounds from the 10 group of peroxides, nitrates, chlorates or perchlorates;
  - c) combustion moderators which are capable of influencing the combustion and its rate by heterogeneous or homogeneous catalysis; and
  - 15 d) optionally also
  - d) additions which are capable of reducing the proportion of the toxic gases.
2. Gas-producing agent according to claim 1, 20 characterised in that it contains as combustibles (nitrogen-containing compounds) one or more tetrazole derivatives of the formula:

25



in which R<sub>1</sub> and R<sub>2</sub> or R<sub>3</sub> can be the same or different, 30 with either R<sub>2</sub> or R<sub>3</sub> being present, and standing for hydrogen, hydroxy, amino, carboxyl, an alkyl radical with 1 to 7 carbon atoms, an alkenyl radical with 2 to 7 carbon atoms, an alkylamino radical with 1 to 10 carbon atoms, an aryl radical, optionally substituted 35 with one or several substituents which can be the same or different and are selected from the amino group, the

nitro group, alkyl radicals with 1 to 4 carbon atoms or an arylamino radical in which the aryl radical can optionally be substituted, or the sodium, potassium and guanidinium salts of the said tetrazole derivatives.

5

3. Gas-producing agent according to claim 2, characterised in that

- R<sub>1</sub> preferably stands for hydrogen, amino, hydroxy, carboxyl, a methyl, ethyl, propyl or isopropyl, butyl, isobutyl or tert-butyl, n-pentyl, n-hexyl, or n-heptyl radical, a methylamino, ethylamino, dimethylamino, n-heptylamino, n-octylamino or n-decylamino radical, a tetrazole radical, a phenylamino radical, a phenyl, nitrophenyl or aminophenyl radical; and
- R<sub>2</sub> or R<sub>3</sub> preferably stands for hydrogen, a methyl or ethyl radical, a phenyl, nitrophenyl or aminophenyl radical.

20 4. Gas-producing agent according to any one of claims 1 to 3, characterised in that

the nitrogen-containing compounds are selected from the group of the tetrazole derivatives and are preferably selected from the compounds 5-aminotetrazole, lithium,

25 sodium, potassium, zinc, magnesium, strontium or calcium 5-aminotetrazolate, 5-aminotetrazole nitrate, sulphate, perchlorate and similar compounds, 1-(4-aminophenyl)-tetrazole, 1-(4-nitrophenyl)-tetrazole, 1-methyl-5-dimethylaminotetrazole, 1-methyl-5-methylaminotetrazole, 1-methyltetrazole, 1-phenyl-5-aminotetrazole, 1-phenyl-5-hydroxytetrazole, 1-phenyltetrazole, 2-ethyl-5-aminotetrazole, 2-methyl-5-aminotetrazole, 2-methyl-5-carboxyltetrazole, 2-methyl-5-methylaminotetrazole, 2-methyltetrazole, 2-phenyltetrazole, 5-(p-tolyl)tetrazole, 5-diallylaminotetrazole, 5-dimethylaminotetrazole, 5-

ethylaminotetrazole, 5-hydroxytetrazole,  
5-methyltetrazole, 5-methylaminotetrazole, 5-n-  
decylaminotetrazole, 5-n-heptylaminotetrazole, 5-n-  
octylaminotetrazole, 5-phenyltetrazole,  
5 5-phenylaminotetrazole or bis-(aminoguanidine)-  
azotetrazole and diguanidinium-5,5'-azotetrazolate, as  
well as 5,5'-bitetrazole and its salts, such as the  
5,5'-bi-1H-tetrazoleammonium compounds.

10 5. Gas-producing agent according to any one of claims  
1 to 4, characterised in that it contains:  
as triazine derivatives, 1,3,5-triazine, as triazole  
derivatives, 1,2,4-triazole-5-one, 3-nitro-1,2,4-  
triazole-5-one, as cyanic acid derivatives, sodium  
15 cyanate, cyanuric acid, cyanuric acid esters, cyanuric  
acid amide (melamine), 1-cyanoguanidine, sodium  
dicyanamide, disodium cyanamide, dicyanodiamidine  
nitrate, dicyanodiamidine sulphate, and as urea  
derivatives biuret, guanidine, nitroguanidine,  
20 guanidine nitrate, aminoguanidine, aminoguanidine  
nitrate, thiourea, triaminoguanidine nitrate,  
aminoguanidine hydrogen carbonate, azodicarbonamide,  
tetracene, semicarbazide nitrate, as well as urethanes,  
ureides such as barbituric acid, and derivatives  
25 thereof.

6. Gas-producing agent according to any one of claims  
1 to 5, characterised in that it contains as oxidants:  
- peroxides of alkali and alkaline earth metals,  
30 zinc peroxide, and peroxodisulphates of the said  
elements and ammonium peroxodisulphate, or  
mixtures of these compounds;  
- ammonium nitrate, nitrates of alkali and alkaline  
earth metals, in particular lithium, sodium or  
35 potassium nitrate, and strontium nitrate, or  
mixtures of these compounds;

- halogen oxycompounds of alkali or alkaline earth metals or of ammonium, preferably potassium perchlorate or ammonium perchlorate, or mixtures of these compounds.

5

7. Gas-producing agent according to any one of claims 1 to 6, characterised in that it contains as oxidant a combination of zinc peroxide, potassium perchlorate and at least one nitrate, preferably sodium nitrate or 10 strontium nitrate.

8. Gas-producing agent according to any one of claims 1 to 7, characterised in that the ratio of the oxidants in the gas-producing mixture is 1:2:10, with a total 15 content of 60% by wt.

9. Gas-producing agent according to any one of claims 1 to 8, characterised in that the ratio of the nitrogen-containing compounds to the oxidants in the 20 mixture is balanced such that on combustion of the gas-producing mixture oxygen is formed in excess.

10. Gas-producing agent according to any one of claims 1 to 9, characterised in that it contains, as 25 combustion moderators, substances or mixtures thereof which are capable of influencing the combustion and its rate by heterogeneous or homogeneous catalysis, the proportion of these substances in the mixture amounting to up to 8%.

30

11. Gas-producing agent according to any one of claims 1 to 10, characterised in that it contains as combustion moderators metals, metal oxides and/or metal carbonates and/metal sulphides or mixtures of these 35 combustion moderators, the metals used preferably being boron, silicon, copper, iron, titanium, zinc or

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molybdenum.

12. Gas-producing agent according to any one of claims  
1 to 11, characterised in that it contains as  
5 combustion moderators sulphur, ferrocene and its  
derivatives.

13. Gas-producing agent according to any one of claims  
1 to 12, characterised in that it contains, as an  
10 addition, substances which are capable of reducing the  
content of the noxious gases nitrogen oxides and/or  
carbon monoxide.

14. Gas-producing agent according to any one of claims  
15 1 to 13, characterised in that it contains as addition  
substance:

- combustion moderators, noble metals such as  
palladium, ruthenium, rhenium, platinum or rhodium  
or oxides of the noble metals, and mixtures of  
20 these compounds, or  
- basically reacting substances such as, for  
example, oxides, hydroxides or carbonates of  
alkali and alkaline earth metals, of zinc, as well  
as mixtures of these compounds, or  
25 - urea, guanidine and derivatives thereof, compounds  
having NH<sub>2</sub> groups such as, for example,  
amidosulphonic acids, amido complexes, amides, and  
mixtures of these compounds.

30 15. Gas-producing agent according to any one of claims  
1 to 14, characterised in that the amount of the  
additions used is about 10 % by wt. in the charge and  
up to 75% by wt. in the outlet passages the amounts  
being based on the gas charge.

35

16. Gas-producing agent for gas generators, comprising

nitrogen-containing compounds, characterised in that it contains:

- a) as nitrogen-containing compound (fuel), a combination of aminotetrazole and the salts, preferably the calcium, magnesium or zinc salts, of aminotetrazole, preferably a combination of 5-aminotetrazole and the corresponding salts of 5-aminotetrazole;
- b) as oxidant, at least three compounds from the group of peroxides, nitrates, chlorates or perchlorates, preferably sodium nitrate and potassium perchlorate; and
- c) combustion moderators which are capable of influencing the combustion and its rate by heterogeneous or homogeneous catalysis, preferably zinc oxide and the carbonates of zinc and calcium.

17. Gas-producing agent for gas generators, comprising nitrogen-containing compounds, characterised in that it contains:

- a) as nitrogen-containing compound (fuel), urea, its salts, its derivatives and their salts, preferably biuret, guanidine, nitroguanidine, guanidine nitrate, aminoguanidine, aminoguanidine nitrate, thiourea, triaminoguanidine nitrate, aminoguanidine hydrogen carbonate, azodicarbonamide, dicyanodiamidine nitrate, dicyanodiamidine sulphate, tetracene and/or semicarbazide nitrate, as well as urethanes, ureides such as barbituric acid, and their derivatives;
- b) as oxidants, at least two compounds from the group of peroxides, nitrates, chlorates or perchlorates, preferably sodium nitrate and potassium perchlorate; and
- c) combustion moderators which are capable of

influencing the combustion and its rate by heterogeneous or homogeneous catalysis, preferably zinc oxide and the carbonates of zinc and calcium.

5 18. Gas-producing agent according to claim 16 or claim 17, characterised in that the oxidants are defined according to claim 6 and the combustion moderators are defined according to any one of claims 10 to 12.

10 19. Method of producing a gas-producing agent for gas generators according to any preceding claim, characterised in that the nitrogen-containing compound or compounds (fuel) is/are mixed with the oxidants, the combustion moderators and optionally with further 15 additions and the mixture is homogenised.

20 20. Method according to claim 20, characterised in that the gas-producing agent is compressed with the use of pressing aids, for example graphite, molybdenum 20 sulphide, Teflon, talc, zinc stearate or boron nitride.

25 21. Method according to claim 20, characterised in that the blanks are coated.

25 22. Method according to any one of claims 19 to 21, characterised in that a definite porosity of the blank is produced for control of the rate of combustion .

30 23. Life-saving system containing a gas-producing agent according to any one of claims 1 to 18.

24. The use of the gas-producing agent according to any one of claims 1 to 18 for the production of gas.

Abstract

The subject of the present invention is gas-producing agents for gas generators which contain, as 5 nitrogen-containing compound (fuel), a compound from the group tetrazole, triazole, triazine, cyanic acid, urea, their derivatives or salts thereof, as oxidants, compounds from the group of the peroxides, nitrates, chlorates or perchlorates, and also combustion 10 moderators which are capable of influencing, by heterogeneous or homogeneous catalysis, the combustion and its rate, as well as, optionally, additions capable of affecting the proportion of the toxic gases.



DECLARATION AND POWER OF ATTORNEY FILED WITH U.S. DESIGNATED OFFICE UNDER 35 U.S.C. 371(c)(4)

As a below named inventor, I/we hereby declare that:

My/Our residence, post office address and citizenship are as stated below next to my/our name, I/we believe that I/we are the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

GAS-PRODUCING MIXTURES

the specification of which was filed as PCT International Application No. PCT/EP96/00605

filed FEBRUARY 13, 1996 and was amended on (if applicable)

I/We hereby state that I/we have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I/We acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56(a).

I/We hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)	Priority Claimed
195 05 568.3 (Number)	GERMANY (Country) 18/FEBRUARY/1995 (Day/Month/Year Filed) <input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
 (Number)	 (Country) (Day/Month/Year Filed) <input type="checkbox"/> Yes <input type="checkbox"/> No
 (Number)	 (Country) (Day/Month/Year Filed) <input type="checkbox"/> Yes <input type="checkbox"/> No
 (Number)	 (Country) (Day/Month/Year Filed) <input type="checkbox"/> Yes <input type="checkbox"/> No
 (Number)	 (Country) (Day/Month/Year Filed) <input type="checkbox"/> Yes <input type="checkbox"/> No
 (Number)	 (Country) (Day/Month/Year Filed) <input type="checkbox"/> Yes <input type="checkbox"/> No

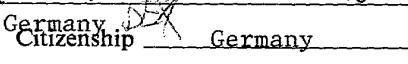
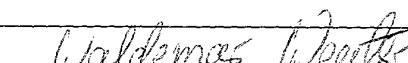
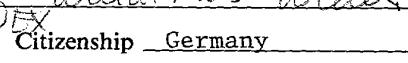
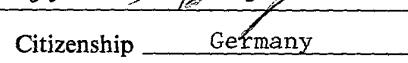
I/We hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I/we acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)	(Filing Date)	(Status: patented, pending, abandoned)
(Application Serial No.)	(Filing Date)	(Status: patented, pending, abandoned)
(Application Serial No.)	(Filing Date)	(Status: patented, pending, abandoned)
(Application Serial No.)	(Filing Date)	(Status: patented, pending, abandoned)

I hereby appoint as principal attorneys: Donald R. Antonelli, Reg. No. 20,296; David T. Terry, Reg. No. 20,178; Melvin Kraus, Reg. No. 22,466; William I. Solomon, Reg. No. 28,565; Gregory E. Montone, Reg. No. 28,141; Ronald J. Shore, Reg. No. 28,577; Donald E. Stout, Reg. No. 26,422; Alan E. Schiavelli, Reg. No. 32,087; James N. Dresser, Reg. No. 22,973 and Carl I. Brundidge, Reg. No. 29,621 to prosecute and transact all business connected with this application and any related United States application and international applications. Please direct all communications to the following address:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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